THIRD YEAR REPORT: Sea-Salt Aerosol in Hawaii and the Influence of Large Organized Structures (LOS) or Rolls on Fluxes and Visibility

Antony D. Clarke
Department of Oceanography
University of Hawaii
1000 Pope Rd.
Honolulu, HI 96822
phone: (808) 956-6215 fax: (808) 956-7112 email: tclarke@soest.hawaii.edu

Steven G. Howell
phone: (808) 956-5185 fax: (808) 956-7112 email: showell@soest.hawaii.edu

Vladimir N. Kapustin
phone: (808) 956-7777 fax: (808) 956-7112 email: kapustin@soest.hawaii.edu

Award Number: N00014-07-1-0031 for period 10/1/2006 – 9/30/2009
http://www.soest.hawaii.edu/HIGEAR/

LONG-TERM GOAL

Our long-term goal is to establish an improved understanding of the factors that control the marine aerosol properties and concentrations as they relate to generation processes, mixing processes, their dependence on oceanic and environmental conditions and physicochemical evolution in the marine boundary layer. We expect these efforts to lead to improved modeling and predictability of marine aerosol concentrations and optical properties.

OBJECTIVES

Our recent ONR efforts characterized sea-salt aerosol (SSA) size distributions and production from breaking waves through measurements under accelerating wind conditions in the "natural wind tunnels" between Hawaiian Islands. These experiments revealed the typical presence of Large Organized Structures (LOS) or rolls aligned along the wind in the channels both with and without visible cloud streets. This organized mixing process can include entrainment of air to/from the buffer layer (BuL), the free troposphere (FT) and the mixed boundary layer (BL) that can directly influence aerosol concentrations, fluxes and visibility. Our activities this past year include analysis of tugboat and aircraft data as part of preparation of our paper investigating these LOS including their role in aerosol processes and the vertical mixing of sea-salt and associated optical extinction.

A second new component of our studies involves deployments of the Aerodyne Time-of-Flight Aerosol Mass Spectrometer (ToF-AMS) to address the evolving issue of organic carbon (OC) aerosol in the marine environment. This OC has been the subject of limited research in the past, but recent papers have argued for large OC sources associated with marine primary productivity. If so, this

Report Documentation Page

Form Approved
OMB No. 0704-0188

1. REPORT DATE	2. REPORT TYPE	3. DATES COVERED
30 SEP 2009	**Annual**	**00-00-2009 to 00-00-2009**

4. TITLE AND SUBTITLE	5a. CONTRACT NUMBER
THIRD YEAR REPORT: Sea-Salt Aerosol In Hawaii And The Influence Of Large Organized Structures (LOS) Or Rolls On Fluxes And Visibility	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **University of Hawaii,Department of Oceanography,1000 Pope Rd.,Honolulu,HI,96822**	8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES
Code 1 only

14. ABSTRACT
Our long-term goal is to establish an improved understanding of the factors that control the marine aerosol properties and concentrations as they relate to generation processes, mixing processes, their dependence on oceanic and environmental conditions and physicochemical evolution in the marine boundary layer. We expect these efforts to lead to improved modeling and predictability of marine aerosol concentrations and optical properties.

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			
unclassified	**unclassified**	**unclassified**	**Same as Report (SAR)**	**6**	

understudied component might impact marine aerosol composition, optical properties and cloud condensation nuclei (CCN).

The AMS is offering an unprecedented ability to explore organic material as a function of particle composition and size. As part of this assessment of interaction of OC with sea-salt, we deployed some of our aerosol instrumentation including our Viasala ceilometer during KA-09-03, a cruise on the NOAA ship Ka`imimoana from August 24 to Sept 24, 2009. We also collaborated with members of the Volkamer group from the University of Colorado, Boulder, who had earlier discovered surprisingly high concentrations of glyoxal over the central Pacific using their MAX-DOAS instrument. The presence of glyoxal, which should be short-lived in the marine boundary layer, implies that some organic material (other than DMS) must be emitted from the ocean and must wind up as organic aerosol. This collaboration also involved evaluating the relation of marine aerosol physical chemistry to measured cloud condensation nuclei (CCN, T. Nenes). Our HR-ToF-AMS is key to understanding the nature of that organic aerosol and related issues.

APPROACH

We focus our ONR efforts upon characterizing the marine aerosol full size distributions, the associated source function and optical properties [*Clarke et al.*, 2003; *Clarke et al.*, 2006; *Clarke and Kapustin*, 2003; *Kapustin et al.*, 2006; *Shinozuka et al.*, 2004]. Our instrumentation includes total and submicrometer nephelometers, particle size spectrometers (DMA, LDMA, OPC, APS), Hot/cold CN, size-resolved volatility (DMA and OPC) and Time of Flight Aerosol Mass Spectrometry (ToF-AMS). The AMS was obtained through an ONR supported DURIP proposal for marine aerosol studies and was deployed this past year on instrumented aircraft and shipboard open ocean experiments. The latter (see Fig. 1 below) was in conjunction with a nephelometer, APS, LDMA, and a cloud condensation nucleus counter (CCN) borrowed from NCAR. Our deck-mounted ceilometer acted as a low-power IR lidar, enabling us to measure boundary layer depth and estimate optical influences of seasalt concentrations. For the first half of the cruise the CCN sampled unmodified inlet air; in the second half, it was replumbed to take air from the outlet of the LDMA. The CCN complemented the rest of the instrumentation it two ways: by identifying the particles most likely to form cloud droplets, and by measuring degree of activation as a function of supersaturation and particle size, which can reveal the presence or absence of organic material at sizes too small for the AMS to effectively sample. Although Ka`imimoana is poorly configured for aerosol sampling and we had little advance notice of the cruise, we were able to obtain valuable new information on marine aerosol even though we were not able to sample the full sea-salt size distribution.

WORK COMPLETED THIS YEAR

Our ONR related efforts this year include the deployment of several of our instruments during a TAO buoy maintenance cruise aboard the NOAA ship Ka`imimoana. The buoys were from 8 N to 8 S along 140 W and 125 W. This time of year the ITCZ is well north of the Equator, so most of the cruise was in apparently clean Southern Hemisphere air. We crossed the ITCZ and sampled Northern Hemisphere air during the transits from and to Hawaii.

Figure 1 shows the overlay of the Navy Operational Global Atmospheric Prediction System (NOGAPS) model for Surface Winds color coded with wind speed (kt) on top of GOES-10 image (NRL Home Page - http://www.nrlmry.navy.mil/). The white line represents the NOAA ship Ka`imimoana track for Julian Days 237 – 248.

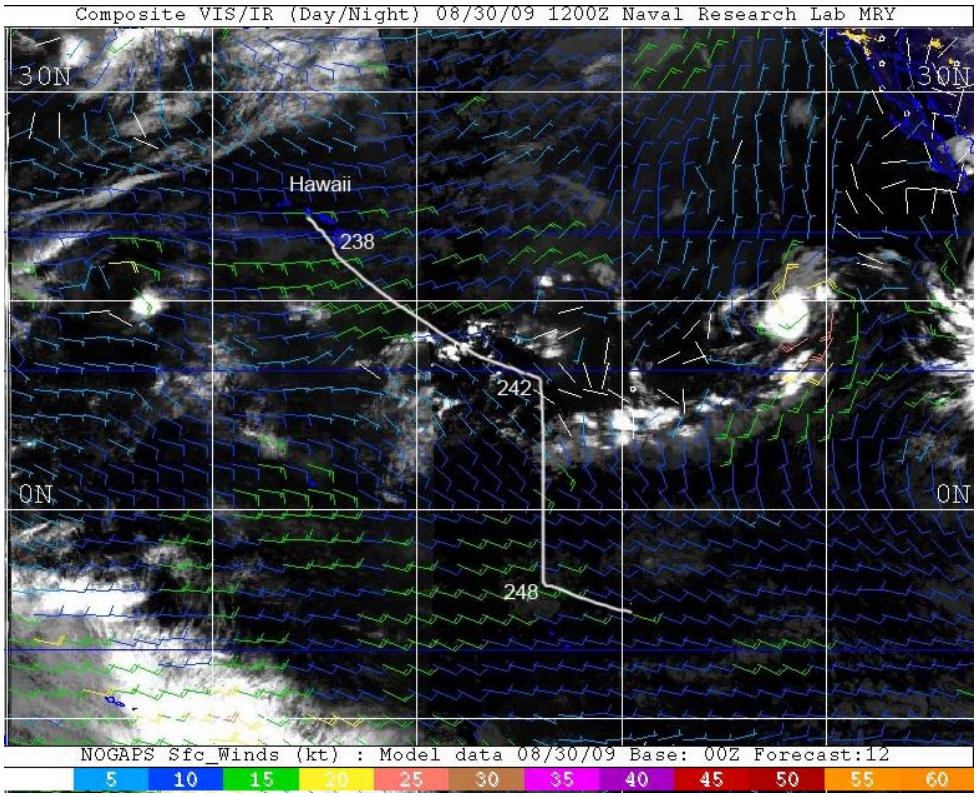

Figure 1. The overlay of the Navy Operational Global Atmospheric Prediction System (NOGAPS) model for Surface Winds color coded with wind speed (kt) on top of GOES-10 image. White line represents the NOAA ship Ka`imimoana track for Julian Days 237 – 248.

NOGAPS output generated every 12 hours (0 and 12 UTC) was interpolated in time to correspond to the current satellite image. While the satellite pictures represent real-time data for 1200Z, JD242 (08/30/09), the model fields are 12h forecasts. Trade winds (green, JD237-241 and JD245-248), ITCZ (white, JD241-242) and low wind regions (blue, JD242-245) are clearly visible.

Since the cruise ended on September 24th, we have only very preliminary results. As shown in Figure 2, the ceilometer and nephelometer results are well correlated but largely independent of AMS mass for sizes smaller then 0.7μm, showing that visibility in the MBL over the central Pacific is governed largely by wind-lofted seasalt rather than photochemically-derived accumulation mode particles or by long-distance transport from the continents. Particularly in the Southern Hemisphere, the accumulation mode was dominated by sulfate, probably derived from dimethylsulfide (DMS). The organic fraction in the Southern Hemisphere was only a few percent and little influenced by productive equatorial upwelling regions. In the Northern Hemisphere gyre (low primary productivity) OC occasionally constituted as much as 30% of the total. Many of the rain events (visible as red streaks to the surface in the ceilometer plot) are accompanied by rapid drops in sulfate, indicating effective scavenging. Interpretation of these observations will be the focus of our efforts in the year ahead.

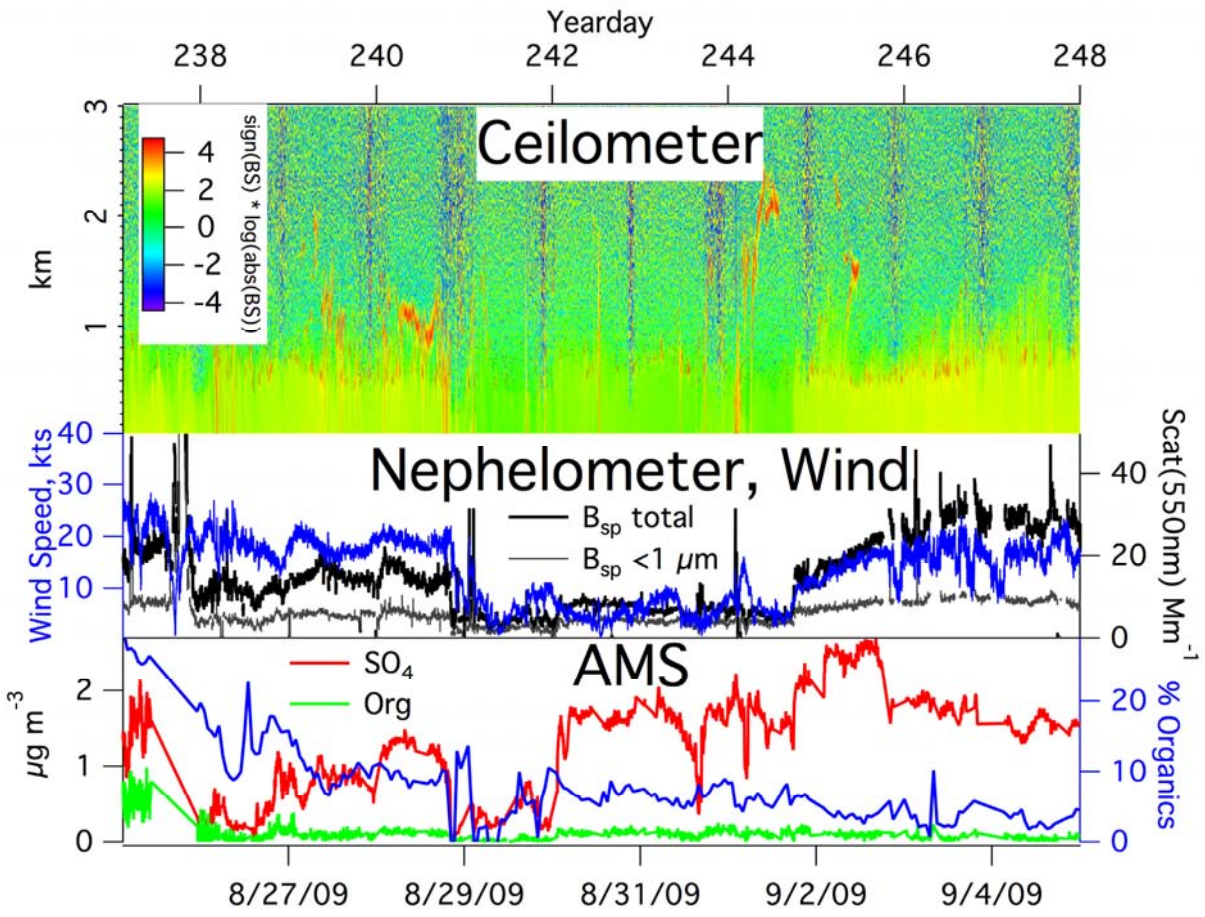

Figure 2. Preliminary data from ceilometer (top panel) – 2D plot of lidar backscattering return, color coded with backscatter intensity; ,nephelometer and wind speed (mid panel) - total(black) and submicron (gray) scattering, wind speed (blue); and AMS (bottom panel) – SO4 (red), Organics (green), Organics fraction (blue).

High-resolution AMS analysis revealed small amounts of organic sulfur, consistent with methanesulfonate, and all of the fragments expected from glyoxal. While most of the organic carbon fragments detected contained just 1 C atom, measurable quantities of longer chain organics were present.

We have also prepared the paper based on our aerosol data measured near Hawaii (tugboat operations) and in equatorial Pacific (the NCAR C-130 as part of PASE experiment). The paper **"Aircraft Observations of Marine Aerosol Properties in the Presence of Boundary Layer Rolls",** *Vladimir Kapustin, Antony Clarke, Steven Howell, Steven Conley, Ian Faloona, Vera Brekhovskikh and Cameron McNaughton,* focused on the direct in-situ marine aerosol properties in the presence of BL rolls (see figure below). We also studied whether the presence of rolls leads to the enhancement of aerosol fluxes.

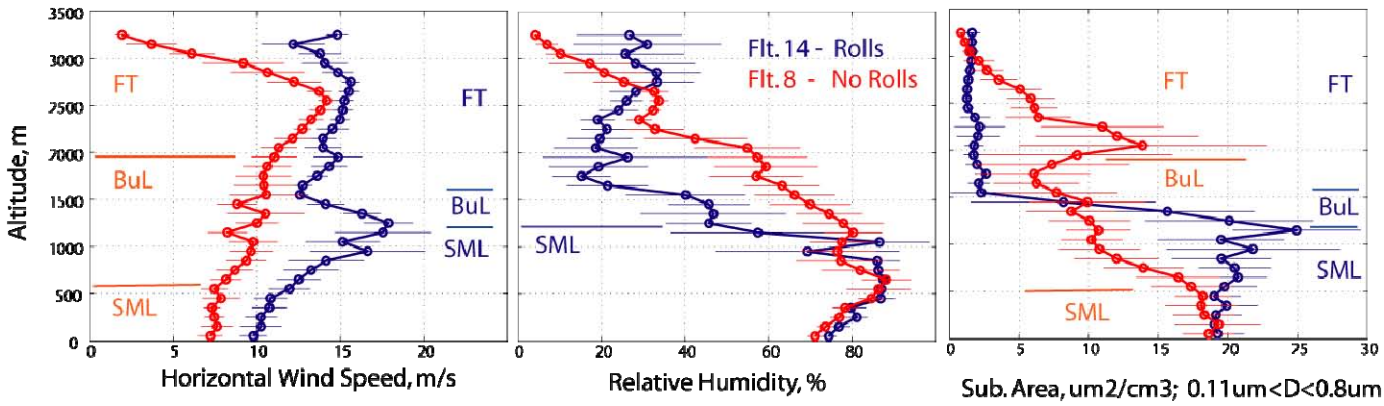

Figure 3. Bin averaged vertical profiles of wind speed (a), relative humidity (b) and submicron aerosol surface area (c) for Flight 14 (blue, numerous rolls present) and Flight 8 (red, no rolls). Horizontal lines indicate the top and bottom of the surface mixed layer (SML), the buffer layer (BuL) and the free troposphere (FT). Higher surface wind speed and wind shear @1200m (left panel) are indicative of BL rolls present (flight 14 –blue). Rolls have a pronounced effect on the multilayer BL structure and aerosol vertical mixing (right panel).

RESULTS

Our preliminary Ka`imimoana cruise results are showing that aerosol scattering and visibility in the MBL over the central Pacific is governed largely by wind-lofted seasalt rather than photochemically-derived accumulation mode particles or by long-distance transport from the continents. Particularly in the Southern Hemisphere, the accumulation mode was dominated by sulfate, probably derived from dimethylsulfide (DMS). The organic fraction in the Southern Hemisphere was quite low (a few percent), while in the Northern Hemisphere it constituted up to 30% of the total. The low OC concentrations and lack of enhancements in regions of elevated productivity raise questions over the large OC fractions recently attributed to ocean biology in other regions (eg. N. Atlantic) that are probably subject to long range transport of OC. High-resolution AMS analysis also revealed small amounts of organic sulfur, consistent with methanesulfonate, and all of the fragments expected from glyoxal, a short-lived organic compound visible to the MAX-DOAS instrument, but at very low concentrations.. While most of the organic carbon fragments detected contained just 1 C atom, measurable quantities of longer chain organics were present. More detailed analysis will follow this coming year.

We are also finalizing the analysis of LOS influence on aerosol evolution in the MBL and preparing related paper for the 2009 submission. Roll structures in the diverse MBL settings demonstrate that these can play an active role in the redistribution of aerosol, gas and water vapor in the MBL compared to non-roll cases. Depending upon the thermodynamic profiles, roll size, altitude and temporal duration, these LOS can have a marked effect on the aerosol exchange between a surface mixed layer, a buffer layer and a free troposphere. This will lead to changes in the optical parameters of these layers relative to regions not influenced by the rolls.

REFERENCES

Clarke, A., V. Kapustin, S. Howell, K. Moore, B. Lienert, S. Masonis, T. Anderson, and D. Covert (2003), Sea-salt size distributions from breaking waves: Implications for marine aerosol production and optical extinction measurements during SEAS, Journal of Atmospheric and Oceanic Technology, 20, (10), 1362-1374.

Clarke, A., S. Owens, and J. Zhou (2006), An ultrafine sea-salt flux from breaking waves: Implications for cloud condensation nuclei in the remote marine atmosphere, J. Geophys. Res., 111, (D06202), doi:10.1029/2005JD006565.

Clarke, A.D., and V.N. Kapustin (2003), The Shoreline Environment Aerosol Study (SEAS): A context for marine aerosol measurements influenced by a coastal environment and long-range transport, Journal of Atmospheric and Oceanic Technology, 20, (10), 1351-1361.

Kapustin, V., A. Clarke, J. Zhou, and V. Brekhovskikh (2006), Sea-salt Aerosol Microphysical, Optical and Spatial Evolution in a Natural Wind-Tunnel, Eos Trans. AGU, 87(36), Ocean Sci. Meeting Suppl., (OS45K-09).

Shinozuka, Y., A.D. Clarke, S.G. Howell, V.N. Kapustin, and B.J. Huebert (2004), Sea-salt vertical profiles over the Southern and tropical Pacific oceans: Microphysics, optical properties, spatial variability, and variations with wind speed, J. Geophys. Res., 109, (D24), 1-18, D24201, doi:10.1029/2004JD004975.

PUBLICATIONS

No new publications in 2009.